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Research in Electrical Phenomena Associated with Aerosols

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QUARTERLY REPORT NO. 4

RESEARCH IN ELECTRICAL PHENOMENA ASSOCIATED WITH AEROSOLS

ABSTRACT

Results of experiments on the charging of particles by spraying them against solid surfaces are presented. Large differences in charging characteristics were exhibited by various liquids. No reasonably consistent charging mechanism is presently available in the literature. Work will continue toward a clarification of the charging process. The effect of charge on the vapor pressure of small particles was evaluated for maximum charging levels. At maximum charge the electrical forces on a droplet are equal and opposite to the surface tension forces and the vapor pressure over a small drop is equal to that over a plane surface. This reduction in vapor pressure is important only for small drop sizes. The deposition of charged particles on small grounded objects was considered. Two geometries were analyzed to show that in spite of low average electric fields there can be a considerable intensification of the field around small grounded objects. As a result charged aerosols will be selectively deposited on these targets. This was demonstrated experimentally.

I. INTRODUCTION

During the period April 15 to July 15, 1961, we continued our study of electrical phenomena associated with aerosols with particular attention to drop charging, the effect of charge on the vapor pressure of small particles, the fate of charge on small volatile particles and the deposition of electrified particles on grounded targets.

II. DEPOSITION OF ELECTRIFIED AEROSOL PARTICLES

It can be shown both theoretically and experimentally that the properties and behavior of aerosols can be considerably modified by electrification. One of the most interesting effects that can be achieved by electrically charging the aerosol particles is to decrease the rate of coalescence and coagulation of the aerosol particles while at the same time increasing their rate of deposition on objects. It appears that techniques for releasing strongly electrified aerosol particles might be potentially useful for increasing the deposition of aerosols on objects and personnel.

The effects of charging the aerosol particles arise directly as the result of electrical forces exerted between the particles and their surroundings. If all of the aerosol particles are given a large charge all of one sign, then a very strong repulsive force exists between the particles that prevents them from colliding with each other.

Laboratory experiments show that aerosol particles can readily be given surface charges in excess of 10^{10} electrons cm^{-2}. It can be shown that when two such drops in the size range of 10 microns approach each other to within one radius the electrical force of repulsion will be several factors of ten greater than the force of gravity. It is clear that because of these large electrical repulsive forces the aerosol particles will not be able to collide and to coalesce or agglomerate.

While the like electrical charge on the charged aerosol particles gives rise to forces that prevent them from coagulating, at the same time it gives rise to forces that can greatly increase the rate of deposition of the particles on surfaces or objects nearby. These forces arise as the result of the equal and opposite electrical charge that each charged particle induces in surrounding objects. With the exception of some clean, dry dielectric substances such as certain plastics the electrical conductivity of most substances such as the earth, rocks, vegetation, the human body and building materials is sufficiently high that an aerosol particle charged with one sign induces an image charge of equal and opposite sign. As a result, when a single charged aerosol particle is near such a surface, it experiences a strong attractive force. When it

comes within a particle radius of such a surface, the attractive force can be a hundred or a thousand times greater than the force of gravity. Accordingly, under such conditions a charged aerosol particle instead of falling downward may move upward and deposit on the under side of a surface.

In addition to the short range image force that acts to precipitate charged aerosol particles there is also a long range precipitation force acting in the same direction.

In the general case in which we are dealing not merely with a single charged aerosol particle but a large mass of such particles each particle will be attracted to the surface not only because of its own induced image charge but because of the image charges induced by all of the other particles. It is clear that since this force depends on the action of all of the particles it will depend strongly on the electrification, concentration, and geometry of the aerosol cloud.

We can obtain some feel for how the magnitude of this precipitating force depends on various factors by considering the problem mathematically for several different simple geometries.

If Q is the charge per aerosol particle, the electrical force acting on each aerosol particle is simply the product of this charge and the electric field F or

$$f = FQ$$

The electric field and hence the force at the surface which the particle approaches is dependent on the size and geometry of the cloud.

Consider the simplest case, that of a horizontal layer of aerosol particles of concentration N per unit volume, and of thickness h over a surface. From Poisson's relation the field at the surface is given by

$$F = 4\pi hnQ$$

Or in words the precipitating force is proportional to the depth, number concentration, and the charge on the aerosol cloud.

It should be recognized that in practice the maximum electric field that can be maintained is limited by the onset of corona discharge from

vegetation, structures or personnel. Over a forested area the maximum field will be about 10 or 20 volts cm^{-1} while over an open field the gradient may be as high as 100 volts cm^{-1}.

Although the average field over the surface of the terrain is limited to rather low values the field will be considerably intensified around exposed objects and personnel and as a result the rate of aerosol deposition here may be far greater than elsewhere and consequently the aerosol will be selectively deposited.

One can make an estimate of the concentration of the field around an object or man by considering the potential in space in which it exists. For example, in a field of 100 volts cm^{-1} a man's head is in a region where the potential is about 20,000 V with respect to ground. Because of the relatively high conductivity of the body the man's head is at ground potential and therefore a corresponding amount of charge has passed from the ground up to his head. If we approximate the head as a sphere of 10 cm radius, its capacity is about 10^{-11} farads so the induced charge on it is about 2×10^{-7} coulombs and the field at its surface is approximately 2000 volts cm^{-1}. Accordingly, we see that the field and hence the rate of aerosol deposition should be about 20 fold greater on the man's head than on the ground.

Another simple and instructive geometrical arrangement that one can consider is the case in which the charged aerosol is on the inside of a sphere. In this case it can be shown that the field on the inside of the sphere arising from the aerosol is given by

$$F = \frac{4}{3} \pi r \, Qn$$

If we integrate to find the potential at the center of the sphere we find that the potential is given by

$$E = \frac{2}{3} \pi r^2 Qn$$

If an object electrically connected to the sphere and small relative to the size of the sphere is placed at its center, the field is concentrated in much the same way as in the previous example and the electrical deposition of particles on the object should be enhanced. In the case where the object is a sphere of radius r' we can calculate that the field F' at

its surface is related to the field at the interior surface of the large sphere by

$$F' = \frac{Fr}{2r'}$$

Experimental verification of this concept was obtained by filling a cubical chamber approximately 1 meter on an edge with a dioctylphthalate (DOP) cloud. A small grounded sphere 3 inches in diameter was located in the center of the chamber. In some cases the aerosol cloud was charged prior to its introduction into the chamber. Corona discharge from fine wires or points was the charging mechanism. Both DC charging and AC charging were used. In both cases, the amount of DOP deposited on the small sphere per unit time for known DOP concentrations was determined. Experiments were run in sets of 2 tests; one test without charging the other test with charging.

Results are given in Table I. Note that considerably more DOP deposited on the target when a charged cloud was used. This was true whether the cloud was charged with AC or DC high voltage. When DC high voltage was used a substantial fraction of the aerosol was lost in the duct work leading into the chamber. Consequently, the concentration of DOP in the chamber was about 1/10 that found when no charging was used. When AC charging was used duct losses were much reduced. So that results could be easily compared, amounts of DOP collected were normalized to the same DOP concentration and these normalized values are reported in Table I.

The fact that deposition was enhanced for both AC charging and DC charging is an interesting one. It has been mentioned earlier that two types of forces affect the deposition, long-range precipitation forces and short-range image forces. Since there is no net space charge in the AC charged cloud, there is no long-range precipitating force and the short-range image force must account for the increased deposition.

The charged clouds exhibited a "target-seeking" capability and from 9 to 83 times as much aerosol deposited on the target relative to that deposited by the uncharged cloud. The spread in the ratio is attributed to differ-

ences in the level of charging and nonhomogenity in the clouds. However, the increases in the amounts deposited are impressive and serve to substantiate the concept discussed above for increasing the deposition of agents on objects and personnel.

TABLE I

RESULTS OF TESTS ON THE DEPOSITION OF ELECTRIFIED PARTICLES ON GROUNDED OBJECTS.

Run No.	Condition of aerosol cloud	Amount of DOP deposited on target *	Ratio: Amount deposited from charged cloud / Amount deposited from uncharged cloud
44-1	uncharged	0.5	49
44-2	charged - AC	24.4	
45-A	uncharged	1.0	9.3
45-B	charged - AC	9.3	
46-1	uncharged	0.7	37
46-2	charged - AC	25.9	
46-1	uncharged	0.7	43
46-3	charged - AC	30	
28-1	uncharged	0.6	50
28-2	charged - DC	30	
28-1	uncharged	0.6	83
28-3	charged - DC	50	
29-4	uncharged	5	15
29-5	charged - DC	75	
29-6	uncharged	4.5	16
29-7	charged - DC	72	
30-1	uncharged	3	35
30-2	charged - DC	105	
31-1	uncharged	4.5	12
31-2	charged - DC	54	

* Normalized to equivalent aerosol concentrations.

III. DROP CHARGING

When a liquid is sprayed from a nozzle the spray is very often found to be electrically charged. When particles are impacted against a target, the deflected particles are often found to be highly charged. These effects are analagous to the common phenomenon known as Triboelectricity. In the latter the relative motion of two solids causes charge separation. Relative motion between a liquid and a gas, a solid and a gas, or a liquid and a solid can also cause charge separation and produce a charged aerosol.

The charging of aerosols by their contact with a solid target is often encountered, particularly when they are being pneumatically transported. Some mechanisms to explain the charging have been suggested but very little agreement among investigators is seen in the literature. Consequently, a program has been initiated to evaluate the magnitude of the contact charging and the influence of physical properties of the test system and to clarify the mechanism which is causing the charge separation.

An apparatus was constructed in which liquids were atomized in an air atomizing nozzle of the painters spray gun type. The spray was directed against a target. The target was grounded through a micro-microammeter. Charging of the aerosol by impaction with the target was seen as an electric current flowing to or from ground through the ammeter. Liquids were sprayed at 10 cc/min. and air pressure was held at 40 psi except where otherwise indicated.

It was recognized that some charging would occur during atomization and all of the charge measured would not be attributable to impact charging. However, a few experiments were made where the charge due to atomizating was evaluated separately. These indicated that charging did occur during spraying but substantially greater charging was produced when the spray was impacted against the target.

Maximum charging was exhibited when the plane of the target was perpendicular to the axis of the spray. The magnitude of the current was dependent upon the spray nozzle-to-target spacing. Results for five different liquids are shown in Figure 1. Note that the currents differ by four orders of magnitude between the dioctylphthalate and kerosene group and the acetonitrile, nitromethane and denimeralized water group. There are two important differences between these groups. DOP and kerosene have low dielectric constants as well as low conductivity. Acetonitrile, nitromethane and denimeralized water have high dielectric constants and are several orders of magnitude more conductive than DOP or kerosene.

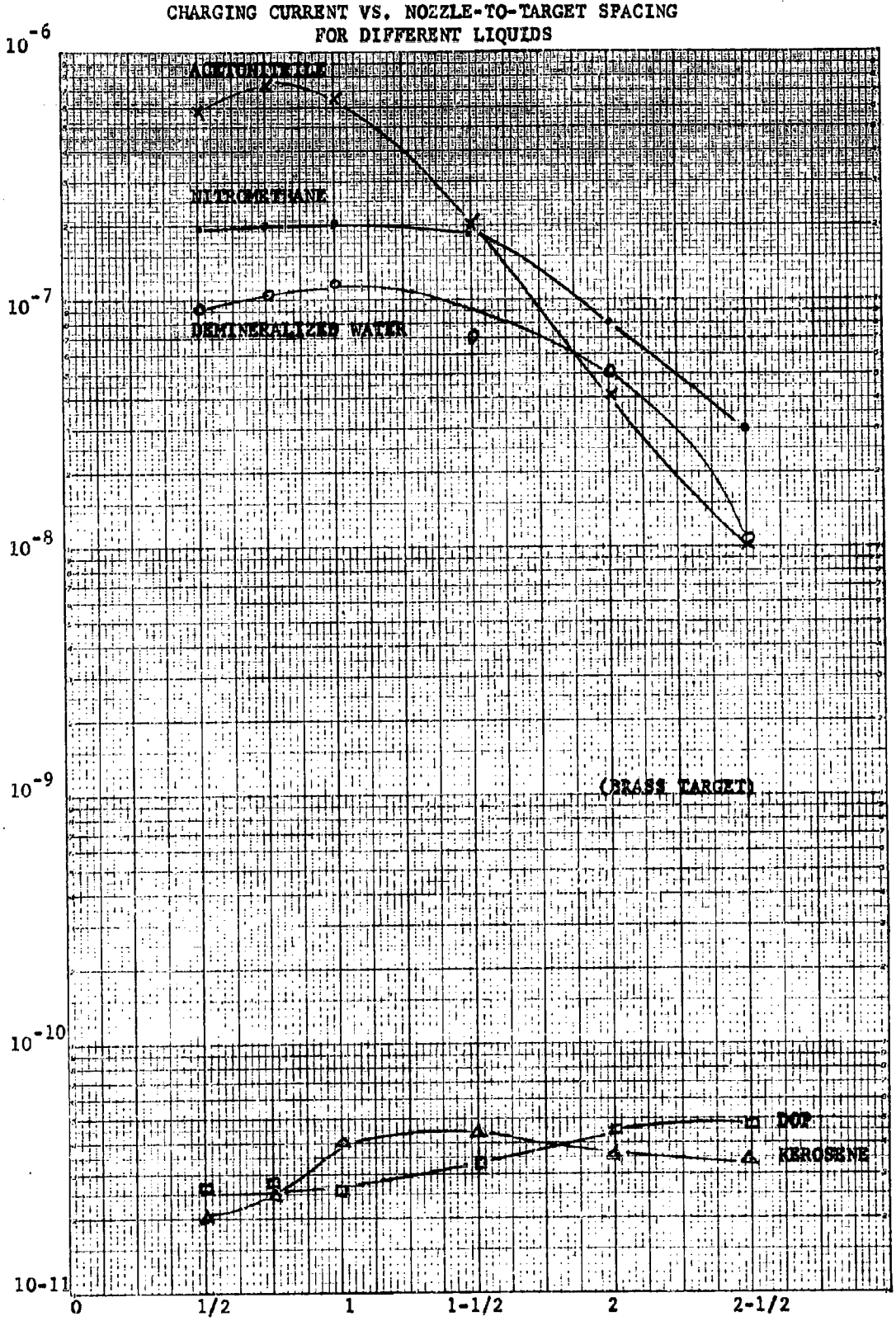
There is another strong indication that conductivity is important. Tap water (high conductivity) gives lower currents than denimeralized water and freshly condensed steam (very low conductivity) gives higher currents than denimeralized water.

The effect of changes in air pressure was of interest. Results are seen in Figure 2, where acetonitrile was impacted against an aluminum target. A wide range of currents was encountered. When flow rate is held constant and air pressure is reduced, larger drops are formed. Reduced charging might be attributed to increased drop size. Unfortunately, drop size is not the only variable which changes with air pressure. The velocity of the air stream varies directly with air pressure. Consequently, a spray droplet produced at lower pressure would have lower kinetic energy. There is additional evidence that kinetic energy is important in the charging process. If we reexamine the upper curves in Figure 1, we note that current falls with increasing nozzle-to-target spacing. With increased spacing the drops would also have lower kinetic energy because of lower velocities caused by aerodynamic drag.

Substitution of various target materials produced changes in charging, sometimes reversing the polarity.

FIGURE 1

CHARGING CURRENT VS. NOZZLE-TO-TARGET SPACING
FOR DIFFERENT LIQUIDS



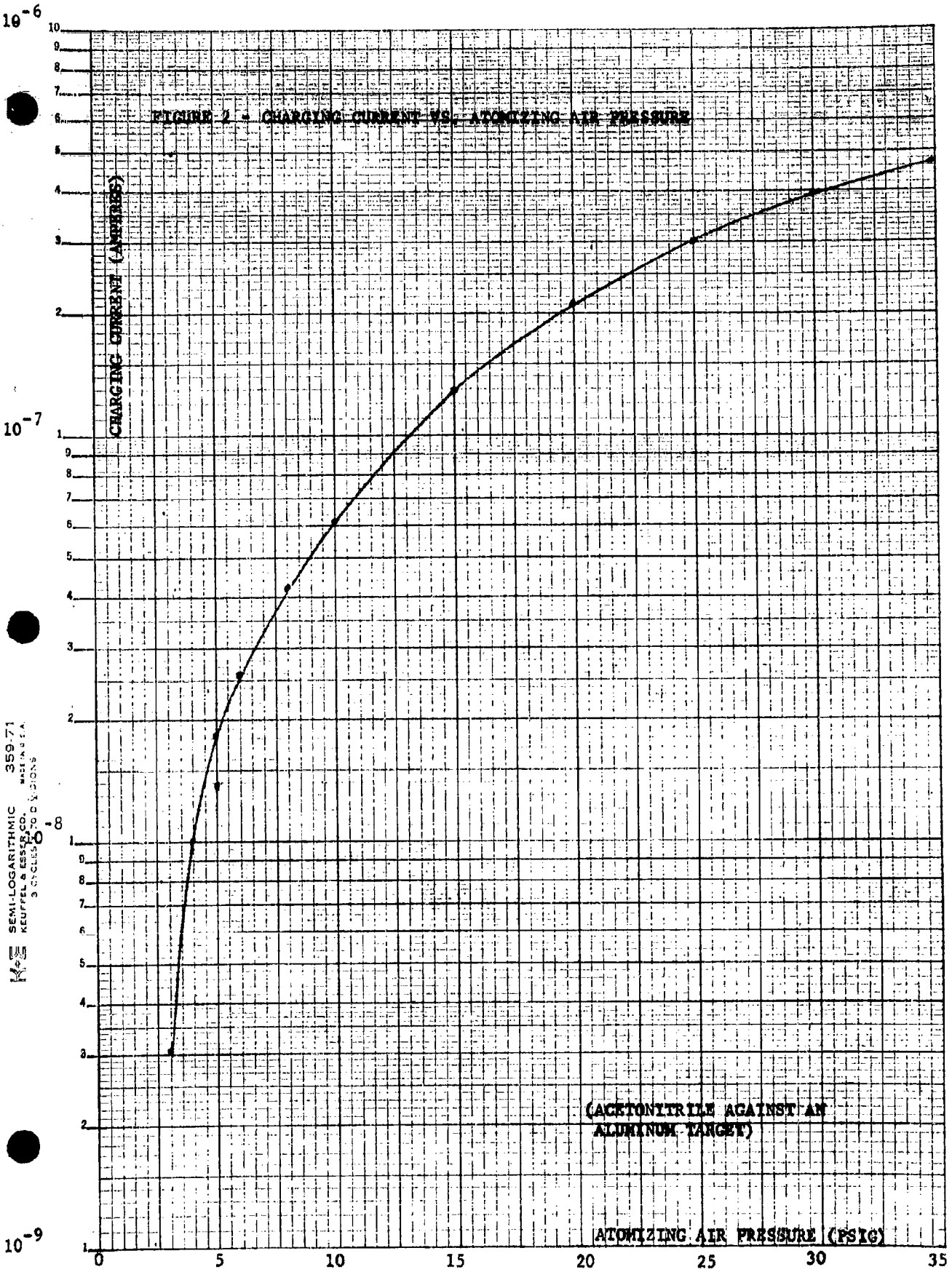


FIGURE 2 - CHARGING CURRENT VS. ATOMIZING AIR PRESSURE
CHARGING CURRENT (AMPERES)
ATOMIZING AIR PRESSURE (PSIG)
(ACETONITRILE AGAINST AN ALUMINUM TARGET)
10^{-6}
10^{-7}
10^{-8}
10^{-9}
0
5
10
15
20
25
30
35
SEMI-LOGARITHMIC 359-71
KEUFFEL & ESSER CO.

We are intrigued with the performance depicted in Figure 1. It is certainly no accident that a difference in current of four orders of magnitude exists. There is some mechanism which would explain this significant difference and it is important that this charging mechanism receive closer study. These first experiments have served well to point out that liquid properties, target properties, geometrical arrangement, etc., are important in the charging process. However, the experiment is far too complex to permit a suitable separation of variables.

Based upon our results and a review of the literature, a second experiment has been devised. A schematic of the apparatus is shown in Figure 3.

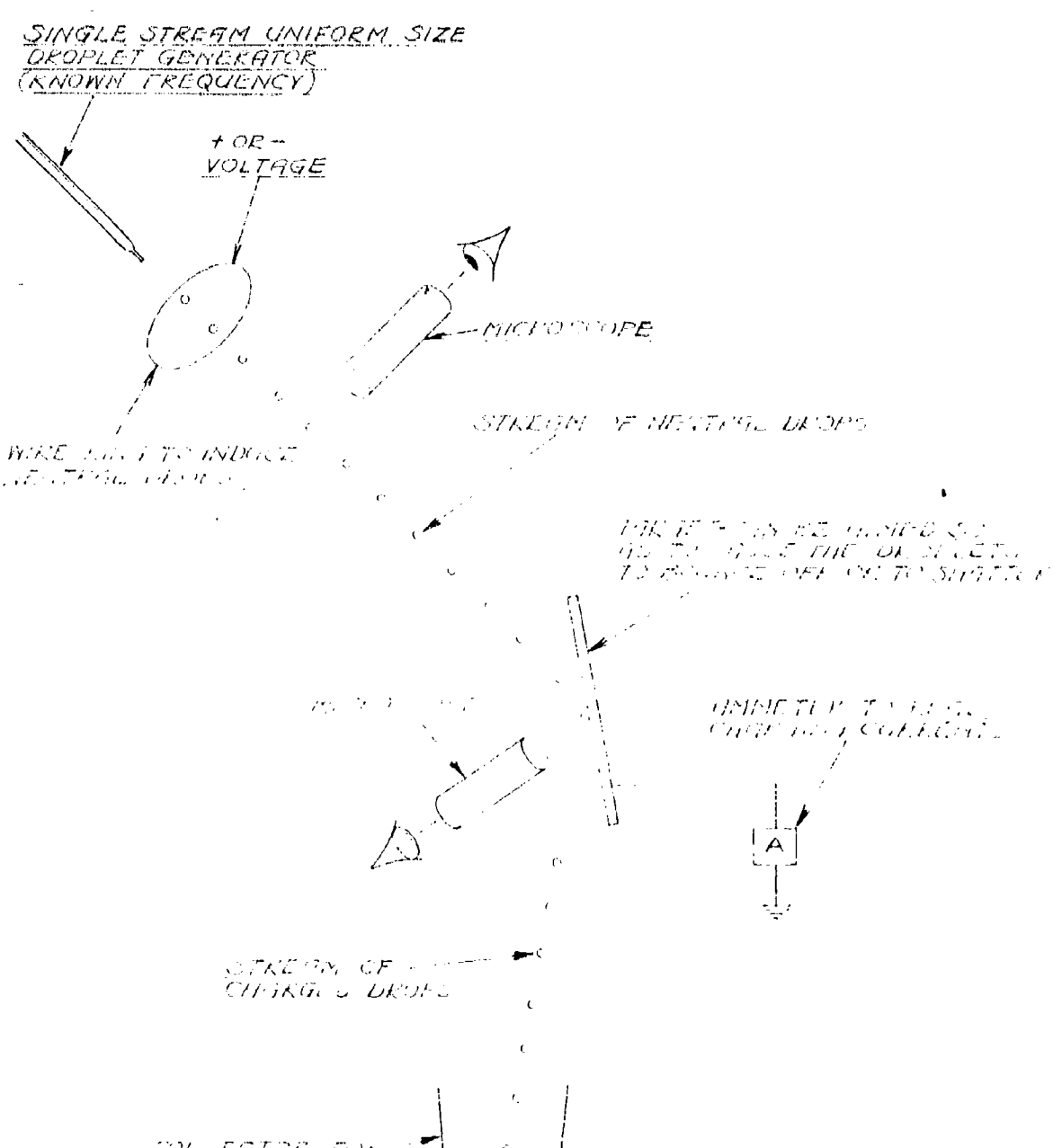
In this arrangement there is no air atomization and, consequently, charging attributable to this factor will be eliminated. Furthermore, it is possible to observe closely the droplet stream and the impact area. This permits accurate size determinations and reveals details of what happens at the time of the impact. The charged ring near the tip of the atomizer will be energized so as to induce neutral drops rather than charged drops which would ordinarily be expected to form. Charging will be evaluated by measuring the charging current or the accumulation of charge in the collector. Charge per drop can then be calculated from the known drop size and frequency.

Various liquids, target materials, drop sizes, impact angles and drop velocities will be studied.

Some experiments have been conducted with this apparatus but without the charged ring near the drop generator. This work indicated that the magnitude of the charge on the particle leaving the dropper was large compared to any charging which may have taken place at the time of impact.

FIGURE 3

APPARATUS FOR THE STUDY OF CONTACT CHARGING



With the charged ring it is possible to reduce the charge on the drops or reverse the polarity of the charge so at some intermediate setting neutral drops must form.

Our review of the literature indicated that there are at present three different theories which explain the charging of drops when they impact on a solid surface. They are (1) a mechanical disruption of the electric double layer at the surface of a drop, (2) inductive charging in the field of the contact potential of the solid surface, and (3) charging due to the existence of fluctuations in the ionic concentration within the drop as it is disrupted by the impaction. The references are cited in Appendix A.

The first theory assumes the existence of a double layer. The outer layer of the charge is, for example, negative and extends from the outer surface to a depth of the order of magnitude of 20 molecular diameters. The positive layer extends to a similar depth below the outer one. These layers have no well-defined boundaries because of the neutralizing effect of osmotic forces. In general, the character of this diffused double layer is not well understood but it is quite sure that it depends heavily on the liquid, the ionic concentration, and the liquid interface (liquid-metal, liquid-air, etc.). The droplet charging is said to result when a larger drop strikes the surface and is broken up. According to the theory, the smaller droplets which are torn from the outer surface of the original drop will contain more of the outer layer and will be charged negatively. The larger droplet will be positive.

The second theory invokes the use of the contact potential of a material. It is well known that at the surface of a solid material there is a potential a few volts different from ground. This potential results in an electric field extending a short distance out from the solid. When

a drop breaks up in the presence of this field, the resulting droplets are said to become inductively charged. This mechanism is clearly closely related to the diffused double layer theory. The field of the contact potential is bound to modify or, in fact, create a diffused double layer in the liquid. The difference, however, is that the double layer theory looks to the solid surface merely as a means of disruption and hence needs no contact potential while the contact potential theory requires no previous separation of ions.

The fluctuational theory is based on the idea that random fluctuations in the ionic concentration within a drop will result in some portions of the drop having a net positive charge and other portions a net negative charge. Thus, when the drop is broken up certain of the droplets will be charged plus and certain minus.

At the present time, none of these mechanisms are well understood and there is a great deal of disagreement as to which of these theories is operative in impact charging. It is possible that all three are operative but then the question is which is dominant. All the three theories are almost wholly qualitative and the data which are available are contradictory.

The paper by Gill and Alfrey[1] supports the contact potential theory and rejects the double-layer theory as an unnecessary complication. In their experiment, they broke up a water drop with an air blast and blew it against the plate of an electrometer. The result was a net charging of the plate which they point out is explained by the contact potential but not by the double layer.

In support of the double-layer theory is the work reported by Guest[2] which shows that in the splashing of water the negative charge comes off as small drops (or possibly ions), while the positive charge comes off as bigger drops. The double-layer theory is also supported by Bikerman[3] and Harper[4].

The paper by Natanson[5] strongly suggests the fluctuational theory. He developed a mathematical model of what the character of the charge should be when using a spray from a bubbler. His experimental results are very close to the theory. Dodd[6] in similar work believes that the double-layer theory can apply only to drops below 10^{-6} centimeters in diameter but not to the drops studies by him and Natanson which are in the low 10^{-4} centimeter range.

The confusion in this field seems to be from lack of good clean experiments and the fact that everyone is trying to explain his own experiments in terms of one theory. The truth may reside in the fact that all three mechanisms are operative but to varying degrees in different experiments. The results of Natanson and Dodd gave little question that the fluctuational theory was a measurable phenomenon, but it cannot explain the work of Gill and Alfrey. Observations of the tendency of big drops to be positive and small ones negative in spraying experiments is difficult to explain except in terms of the double-layer theory.

The conclusion that can be drawn from all of this is that more work is needed.

IV. CHARGE ON A VOLATILE PARTICLE

Two experimental approaches were made in an attempt to determine the fate of charge on a volatile drop. In one case a charged droplet was located on an insulating fiber pendulum in an electric field. The goal was to observe the drop while it evaporated and compute the charge at any time from the displacement of the pendulum. In practice the charge on the pendulum itself was found to dominate the experimental results so as to obscure the effect of the drop charge on pendulum displacement.

An alternative experiment was devised where charged water drops were allowed to fall through air. The initial charge and mass of the drops were measured and the charge and mass at the conclusion of their fall were measured. Because of their charge the falling drops dispersed considerably and spread over a wide area on the collecting surface. Since it was necessary to collect all of the drops it was required that their trojectory be kept reasonably short to minimize the dispersion. This dictated that the time interval be short. Several sets of measurements were made but it was not possible to achieve a time interval long enough to really see what was happening.

Preliminary experiments indicate that a third technique has an excellent chance of achieving success. This is one where a charged drop is suspended in an electric field in a Millikan oil drop type experiment. The movement of the drop in the field is observed. By proper control of the polarity and magnitude of the field a given drop can be caused to hover for extended periods. It can be observed while it evaporates. Drop charge can be determined if one observes the electric field, drop size and drop density.

V. EFFECT OF CHARGE ON THE VAPOR PRESSURE OF SMALL LIQUID DROPLETS

The surface tension of a liquid droplet generates a compressive force which raises the pressure on the concave side of a liquid surface according to the relationship

$$p = \frac{2\gamma}{r}$$

where p is the excess pressure in the liquid droplet, γ is the surface tension and r is the droplet radius.

This excess pressure gives rise to an increase in the equilibrium vapor pressure of the liquid. For small droplets where the excess pressure is greatest, the largest increase in vapor pressure above that for a plane surface is achieved. The increase in vapor pressure is expressed by the following relationship

$$\ln \frac{P}{P_o} = \frac{2\gamma}{r} \left(\frac{M}{\rho RT}\right)$$

where P_o is the vapor pressure over a plane surface, P is the vapor pressure over the curved surface, ρ is the density of the liquid, M is the molecular weight of the liquid, T is the absolute temperature, and R is the gas constant.

If the droplet possesses a surface charge, electrical pressures must be considered and the equation is no longer valid. The term $(\frac{2\gamma}{r})$ must be modified to reflect the electrical pressure which is given by the following relationship:

$$p_e = \frac{F^2}{8\pi}$$

where p_e is the electrical pressure and F is the electric field at the

surface. Since this pressure is directed radially outward, it must be subtracted from the surface tension pressure term. The corrected form of the equation for the case of a charged droplet is:

$$\ln \frac{P}{P_o} = \left(\frac{2\gamma}{r} - \frac{F^2}{8\pi}\right)\left(\frac{M}{\rho RT}\right)$$

The magnitude of the terms of equation are given in Table II for water drops of various sizes. The electrical term $(\frac{F^2}{8\pi})$ is evaluated for a potential gradient of 30,000 volts per centimeter which is the usually stated breakdown value for air. It is seen that the electrical term is two orders of magnitude less than the surface tension term for a 75μ diameter drop and several orders of magnitude less for smaller drops. One might hastily conclude that the electrical effect is of little consequence. This is definitely not the case.

We have found in our work that much higher potential gradients occur than that stated maximum. For example, a 37.5 micron (radius) water droplet can be charged until there is an electric field at its surface of 300,000 volts per centimeter. For this condition the electrical pressure term becomes 4×10^4. Note that this is exactly equivalent to the surface tension pressure term given in Table II for this size drop. A water drop of 50 micron radius can be charged until the electric field at the surface is 275,000 volts/cm. The electrical pressure term in this case is also just equivalent to the surface tension term. Similarly a 10 micron radius DOP drop can be charged to 425,000 volts per cm which also equals the excess pressure in the drop due to surface tension.

TABLE II

EFFECT OF SURFACE TENSION AND CHARGE ON THE VAPOR PRESSURE OVER LIQUID DROPLETS

(Calculated and Experimental)

Drop Radius (microns)	$\frac{2\gamma}{r}$	$\frac{F^2}{8\pi}$ (calculated based upon 30,000 volts/cm or 100 ESV/cm)	$\ln \frac{p}{p_o}$	$\frac{p}{p_o}$
0.01	1.4×10^8	4×10^2	0.1	1.15
0.1	1.4×10^7	4×10^2	0.01	1.01
1.0	1.4×10^6	4×10^2	0.001	1.001
10.0	1.4×10^5	4×10^2	0.0001	1.0001
37.5	4×10^4	4×10^2	0.00003	1.00003
50.0	3×10^4	4×10^2	0.00002	1.00002
37.5	4×10^4	4×10^4 (experimental)	0	1
50.0	3×10^4	3×10^4 (experimental)	0	1

Note in Table II that for large drops the surface tension term is small and the vapor pressure over the liquid drop is essentially the same as over a plane surface. Since the electrical term cannot get larger than the surface tension term, the vapor pressure cannot be made lower than that over a plane surface.

There is no doubt that vapor pressure of small drops can be reduced by charging the drops. In the limiting case the equilibrium vapor pressure of the charged drop is exactly equal to the equilibrium vapor pressure over the plane surface. The extent of the effect depends entirely upon the properties of the liquid in question. Since surface tension and molecular weight are subject to wide variations between liquids, the vapor pressure increase over a curved surface is also variable. Those liquids which show large increases in vapor pressure will show the largest reduction in vapor pressure when charged.

VI. FUTURE WORK

During the next quarter we plan to continue studies on the charging of aerosols. The revised experimental technique of Figure 3 will be utilized to study impact charging. We will look at such variables as particle size, liquid properties, target materials, and contact angle.

We will continue to pursue the problem of charge on a volatile particle. We plan to use a Millikan oil drop type experiment, where charged volatile drops will be held in space for extended periods of time by the use of electrostatic fields. With this technique we hope to determine what happens to the charge on a volatile drop.

Experiments with the behavior of water drops show that whether or not two colliding drops coalesce depends very much on the electrical charge on the water drops and on the electric field. It is probably that aerosol particles of organic liquids behave in a similar way. Studies on the coagulation or bounce-off of two drops are of interest in that they are applicable to the stability of an aerosol cloud. We plan to initiate work on the coagulation of charged particles during the next quarter.

APPENDIX A

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Arthur D. Little, Inc.
Cambridge, Massachusetts

ELECTRICAL PHENOMENA ASSOCIATED WITH AEROSOLS - Vonnegut et al.

Report (Quarterly) No. 4, 15 July 1961, 24 pp - 3 illus
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Arthur D. Little, Inc.
Cambridge, Massachusetts

ELECTRICAL PHENOMENA ASSOCIATED WITH AEROSOLS - Vonnegut et al.

Report (Quarterly) No. 4, 15 July 1961, 24 pp - 3 illus
Contract DA18-108-405-Cml-852
CP-0-405-16999

Results of experiments on the charging of particles by spraying them against solid surfaces are presented. Large differences in charging characteristics were exhibited by various liquids. No reasonably consistent charging mechanism is presently available in the literature. Work will continue toward a clarification of the charging process. The effect of charge on the vapor pressure of small particles was evaluated for maximum charging levels. At maximum charge the electrical forces on a droplet are equal and opposite to the surface tension forces and the vapor pressure over a small drop is equal to that over a plane surface. This reduction in vapor pressure is important only for small drop sizes. The deposition of charged particles on small grounded objects was considered. Two geometries were analyzed to show that in spite of low average electric fields there can be a considerable intensification of the field around small grounded objects. As a result charged aerosols will be selectively deposited on these targets. This was demonstrated experimentally.

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